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NUCLEAR CHEMISTRY RESEARCH

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I. INTRODUCTION

In 1974 the charge-state program (Section A, below) was directed toward heavier ions than had been studied previously in our laboratory, largely as a result of the installation of a new ion source at the Van de Graaff (Section G).

The low-level counting program was continued in two projects: a search for natural alpha radioactivity in osmium (Section B) and a search for strontium activity in marine mollusc shells (Section C). In parallel with the latter work, a comprehensive literature review was made of the applications of radiochemistry to oceanographic research (Section D).

A new, non-experimental project was begun: a general, consistent mathematical analysis of low-level counting statistics (Section E). Our computational capabilities were also expanded during the year (Section F).

Finally, we report on the substantial improvements in facilities and equipment which were associated with our move into the University's new chemistry building (Section G).

II. SUMMARY OF RESEARCH

A. Charge-State Distributions of Heavy Ions

1. Equilibrium Charges of Nickel Ions.

This work has been undertaken during the past year for two major purposes: to obtain equilibrium charge-state data on heavy ions not previously available, and to study shell effects in the stripping of heavy ions. Nickel ions were chosen as the first system to study because of their availability from the new Van de Graaff source, because no significant data currently exist on the charges of nickel ions, and because the range of nickel ion energies available at the Van de Graaff can provide information about the crossing of the M-N shell juncture in the interaction of the ions' electrons with the stripping medium. Data are therefore being collected on the equilibrium charge-state distributions of these ions as a function of energy.

Equilibrium charge-state distributions are usually ... symmetrical with respect to charge, although in certain energy regions asymmetries have been observed. Moak, et al. [Phys. Rev. Letters 18, 41 (1967)] reported distortions in the distributions of bromine and iodine ions. Bromine ions passing through solids, for example, give nearly Gaussian distributions at lower energies, but at 140 MeV where L-shell electrons are being removed, the distribution becomes distorted, the lower charge-state fractions being enhanced. This is presumably

because at charge 25 all the M-shell electrons have been removed and higher charge-states can only be achieved by the removal of L electrons. Since the L electrons are more tightly bound than the M electrons, it is more difficult to reach the higher charge-states, whose abundances are therefore suppressed. Similar effects have subsequently been noted in bromine and iodine ions near the M-N and N-O shell junctures by Ryding et al. [Phys. Rev. 185, 129 (1969)] and by Wittkower and Ryding [Phys. Rev. A 4, 226 (1971)].

Moak, et al. also reported apparent shell effects in ^{127}I ions in carbon. When the charge-state fractions are plotted against energy, the energy spacing between the curves increases at charge 25, where the N shell is empty and electrons must be removed from the M shell to form the higher charge-states. In addition, the peaks in the curves become higher as the N shell is emptied.

In earlier work in our laboratory [Phys. Rev. 17, 30 (1968)], we examined the effects of shell structure in highly stripped ions of ^{10}B , ^{12}C and ^{13}C in plastic films and in carbon. Attention was focused on the velocity at which two charge-state fractions F_i and F_{i-1} are equal, which was presumed to correspond to the condition in which the capture and loss cross sections of an electron in the ion are equal. When the ratio F_i/F_{i-1} was plotted against velocity, the velocity spacing between the lines was found to increase at that velocity at which the L shell had been emptied. For electrons within a shell, the velocity spacings were found to be approximately constant.

Such information on shell effects may be valuable in the

development of theories of energy loss and of the capture and loss of electrons in matter. However, the data are not readily extrapolated to other ions.

In the present work, several different ions are being studied over a range of energies in order to obtain charge-state data corresponding to the stripping of electrons from various electronic shells.

A beam of ^{58}Ni ions incident on carbon foils was chosen to initiate this work. In these experiments, the typical output of ^{58}Ni negative ions from the source is of the order of 500 na. Losses during acceleration usually reduce this intensity to approximately 50 na. After the beam is analyzed and passes through the various slit systems, it is reduced to about 10^9 particles/sec at the target. The beam current must be kept extremely low to protect the solid-state detectors, which are susceptible to damage from heavy ions, and to minimize damage to the target foil.

The ^{58}Ni ions emerging from the carbon foil are observed at 0° to the incident beam and analyzed for charge and energy in the Engle split-pole spectrograph. The ions are detected on the focal plane of the spectrograph by two 7 X 45 mm position-sensitive detectors, which permit one to correct for beam fluctuations by counting two charge states simultaneously. The major advantage of using this technique instead of using an electrostatic analyzer for the simultaneous detection of all charge states is that very small charge fractions can be measured with high precision without exposing the detector to excessively high counting rates from the more abundant

charge states. Largely for this reason, design work on the electrostatic analyzer described in last year's report has not been pursued further.

The signals from the detectors are amplified and shaped, and then undergo pulse-height analysis. The data obtained in this way are either stored on magnetic tape or transferred directly to the computer for immediate analysis.

Although the negative ion source is still in the testing stage at the Van de Graaff, preliminary charge-state distributions for ^{58}Ni ions at energies ranging from 21 to 40 MeV have already been obtained. These data are shown in Table I and are plotted as a function of the ion's energy in Figure 1. The mean charge $\bar{q}$, the distribution width d , and the skewness of the distribution s are also included in Table I. The width of a charge-state distribution is defined as

$$d = [\sum_{q'} (q' - \bar{q})^2 F(q')]^{1/2},$$

where $F(q')$ is the fractional abundance of the charge-state q' .

The skewness parameter s is defined as

$$s = \sum_{q'} (q' - \bar{q})^3 F(q') / d^3.$$

As expected, the mean charge of the distributions increases with increasing beam energy. No previously-published data exist for Ni ions at these energies for comparison. However, one can compare these results with values obtained using semi-empirical expressions for the mean charges of ions passing through solids. Nikolaev and Dmitriev [Phys. Lett. 28A, 277 (1968)] give an

FIG. 1. CHARGE-STATE ABUNDANCES OF ^{58}Ni IONS EQUILIBRATED IN
20 $\mu\text{g}/\text{cm}^2$ OF CARBON

Table I. Charge-States of Nickel-58 Ions on Emerging from a 20 $\mu\text{g}/\text{cm}^2$ Carbon Foil

INCIDENT ENERGY (MeV)	CHARGE STATE	% Abundance	
21	8+	0.056	
	9+	0.554	
	10+	3.407	
	11+	11.436	
	12+	22.543	$\bar{q} = 13.00$
	13+	25.839	$d = 1.51$
	14+	20.156	$s = 0.14$
	15+	10.908	
	16+	3.983	
	17+	0.971	
	18+	0.147	
30	9+	0.040	
	10+	0.376	
	11+	2.311	
	12+	7.846	
	13+	16.224	$\bar{q} = 14.52$
	14+	22.595	$d = 1.62$
	15+	22.845	$s = -0.016$
	16+	16.448	
	17+	8.293	
	18+	2.799	
	19+	0.213	
	20+	0.011	
40	10+	0.050	
	11+	0.398	
	12+	2.003	
	13+	6.239	
	14+	12.963	
	15+	20.271	$\bar{q} = 15.61$
	16+	22.546	$d = 1.62$
	17+	19.885	$s = 0.041$
	18+	12.223	
	19+	2.159	
	20+	0.248	
	21+	0.016	

expression for the mean charge:

$$\bar{q}/Z = [1 + (Z^{-\alpha} V/V')^{-1/k}]^{-k}$$

where $V' = 3.6 \times 10^8$ cm/sec, $\alpha = 0.45$ and $k = 0.6$. Using this relation, values of $\bar{q}$ for ^{58}Ni ions passing through a solid at 21, 30 and 40 MeV are 13.5, 15.2 and 16.6, respectively. The present data are within one unit of charge of these values, which is the expected extent of agreement with this expression.

The widths of charge-state distributions are generally found to be very nearly independent of ion velocity, and such appears to be the case for our Ni data.

Semi-empirical relations developed by Dmitriev and Nikolaev [Sov. Phys. JETP 20, 409 (1965).]:

$$d = d_1 Z^w,$$

where $d_1 = 0.38$ and $w = 0.40$,

and by Betz et al. [Phys. Lett. 22, 643 (1966)]:

$$d = 0.27 Z^{1/2},$$

give widths of 1.44 and 1.43, respectively, for Ni ions in Both values are lower than observed in the present work. Another expression developed by Nikolaev and Dmitriev [Phys. Lett. 26A, 277, (1968)]:

$$d = d_1 \{ \bar{q} [1 - (\bar{q}/Z)^{1/k}] \}^{1/2},$$

where $d_1 = 0.5$ and $k = 0.6$, gives 1.53, 1.55 and 1.56 at 21, 30 and 40 MeV, respectively.

Equilibrium charge-state distributions are frequently

assumed to be approximately Gaussian. Although this has been found not to be true in several cases, the distributions we obtain for Ni ions are quite symmetrical, as evidenced by the small values of skewness shown in Table I. It has been generally observed that solid strippers produce more symmetrical charge-state distributions than do gases of comparable mass.

In many cases, ions in the distribution with charges greater than the mean charge show a significantly slower decrease of abundance with energy than do the lower-charged ions. This is seen in our distributions at 21 and 40 MeV, but the distribution at 30 MeV has a negative skewness, indicating that the abundances of low charge-state ions, rather than high charge-state ions, are enhanced.

The energy range of Ni ion measurements is to be expanded, to provide data near the shell transitions. In addition, experiments are to be carried out on other ions in order to compare the effects of different shells.

2. Recoils from the $^{12}\text{C}(^{16}\text{O}, \alpha)^{24}\text{Mg}$ Reaction

The $^{12}\text{C}(^{16}\text{O}, \alpha)^{24}\text{Mg}$ study reported upon last year was continued, but in the course of the work the cross section for the production of ^{24}Mg recoils at 10° to the incident beam was found to be prohibitively small. The intensities of individual ^{24}Mg peaks were found to be comparable to background.

Peak identification was begun by doing coincidence counting of the ^{24}Mg recoils and the alpha particles. A collimated surface barrier alpha detector was placed in the

scattering chamber at angles corresponding to ^{24}Mg recoils at 10° . The angles at which the detectors were placed were chosen also to minimize interference from light reaction products, including alpha particles from other reactions. Thin absorber foils were placed in front of the alpha detector to stop heavy-ion reaction products and scattered oxygen ions. Electrostatic plates, biased at 1000 V and placed in front of the detector swept out electrons. The ^{24}Mg recoil detector was located on the focal plane of the spectrograph. Coincident events were analyzed in the two-dimensional mode of a multi-channel pulse height analyzer. The ^{24}Mg energy information from the position-sensitive detector was stored versus the α energy spectrum from the surface-barrier detector.

Since the objective of this experiment was to observe pre-equilibrium charge states of the ^{24}Mg recoils, extremely thin carbon targets were of course needed. Because of this and because of the stringent coincidence requirements which had to be imposed, both of which decreased the counting rates to impractical levels, this experiment was discontinued in favor of using our Van de Graaff time in more productive ways.

B. A Search for Natural Radioactivity in Osmium

The search for alpha instability in naturally-occurring ^{184}Os , which had just been begun at the time of last year's report, has been completed.

As was reported last year, it appeared that ^{184}Os might have a detectable alpha half-life, based upon alpha-decay theory and energetics. The Low Beta II low-level proportional counter was installed in our new counting room, and an isotopically-enriched sample of ^{184}Os was subjected to chemical processing and an extended period of counting. The result was that, within statistical uncertainties, no activity was observed, but that the lower limit for the half-life of ^{184}Os could be set at a substantially higher value than had previously been possible.

The ^{184}Os sample was obtained as metal powder, enriched from the normal abundance of 0.02 atom percent to 2.25 atom percent. Its isotopic analysis is shown in Table II. A counting sample was prepared by electrodepositing Os metal onto a silver foil disc from a solution of an anionic complex of OsO_4 and sulfamic acid. Experimentation showed that the required complex could best be formed by absorbing gaseous OsO_4 in an aqueous solution containing a forty-fold (molar) excess of sulfamic acid.

Osmium tetroxide (b.p. 130°C) was formed by heating a 17mg sample of enriched osmium metal powder at 200°C for two hours in a stream of pure oxygen gas. The gas was collected in the sulfamic

Table II. Isotopic Analysis of ^{184}Os Sample

<u>Isotope</u>	<u>Atom Percent</u>	<u>Precision</u>
184	2.25	0.05
186	4.79	0.05
187	2.52	0.05
188	16.9	0.1
189	16.8	0.1
190	24.2	0.2
192	32.5	0.3

acid solution at 0°C. The solution was then stored, covered, at room temperature for several weeks, a procedure which was found to increase the chemical yield of the subsequent electrodeposition. Since OsO_4 is reduced by most organic substances, both the stop-cock grease in the distillation apparatus and the covering on the beaker during the storage period had to be of non-oxidizable inorganic materials.

The solution was made alkaline by the addition of 1 ml of 12 M NaOH, then diluted to 50 ml and transferred to an electrodeposition cell. The cell's anode was a 2.5-cm diameter disc of 1 mil Pt foil suspended horizontally, 2.5 cm above and parallel to the cathode. The latter was a weighed disc of 99.9% pure, 2-mil Ag foil, 4.76 cm in diameter, its bottom face spray-coated with Teflon to prevent the deposition of osmium. The electrolysis was carried out at 65 - 75°C for two hours at a constant current density of 2.8 ma/cm², following which the cathode was removed, cleaned of its Teflon coating, rinsed, dried and weighed. The result was a smooth, bright metallic deposit of enriched osmium weighing 6.2 mg and 0.35 mg/cm² in thickness.

A sample of 16.2 mg of 99.999% powdered osmium metal of natural isotopic composition was distilled and electrodeposited identically, producing a 9.0 mg "blank" counting sample, 0.50 mg/cm² in thickness.

The counting was performed on the Beckman Low Beta II gas-flow proportional counter with automatic sample changer. The detector, 5.7 cm in diameter and fitted with an 80 µg/cm² Mylar

window, was shielded by a 15 cm X 15 cm anticoincidence cosmic ray guard detector and by 10 cm of low-activity lead in all directions. Background on the alpha-proportional plateau at 1225V was 0.05 cpm. Beckman P-10 counting gas from a single production batch was used throughout the 5-week counting period. Whenever a new tank of gas was installed, the plateau curve was rechecked before the resumption of counting.

The pulses from the amplification circuitry of the Low Beta II were fed to a Packard Spectrazoom 200-channel multichannel analyzer. A 3.14-MeV alpha peak, obtained by degrading ^{210}Po alpha particles with 2.52 mg/cm² of Mylar, was used to calibrate the analyzer's energy scale.

The two osmium samples were cycled alternately into counting position for periods of time averaging 840 min. The sample-changing routine was scheduled so that the counting periods of the two samples were equally distributed between night and day, weekdays and weekends. Each sample was counted for a total of 25,000 min.

When a sample is counted for a fixed time to determine its radioactivity, the raw data consist of a gross number of counts x observed in a time t and a number of background counts y observed in a time s . The "true" average counting rate of the radioactive sample, γ , is then estimated as

$$\hat{\gamma} = x/t - y/s.$$

The raw counting data shown in Table III give a net counting rate estimate of $\hat{\gamma} = 0.00056$ cpm.

Table III. Counting Data for ^{184}Os

<u>Sample</u>	<u>Number of Counts</u>	<u>Minutes Counted</u>
^{184}Os Enriched	1,259	25,000
Natural Osmium	1,245	25,000

The question of with what probability this result indicates a positive net activity for the ^{184}Os sample (i.e., that $\gamma > 0$) can be answered by applying statistical theory to the case of a background-dominant counting experiment, as has been done by Nicholson [W. L. Nicholson, *Nucleonics* 23, 118 (1966)]. Since the present data satisfy Nicholson's criteria for the valid application of normal distribution theory, that theory's null-hypothesis testing rule was applied to the data. The result is that $\gamma = 0$ with 95 percent certainty. Thus, it is concluded that alpha activity from

^{184}Os has not been observed. This conclusion is verified by the spectral data of Fig. 2, which show no apparent differences between the sample and blank in the energy region of the expected ^{184}Os peak.

To extract a lower limit on the half-life from these data, the counting efficiency must be known. This was determined by counting two 0.0025 μCi sources of ^{210}Po , prepared in the same geometry as the osmium samples and calibrated with a solid-state detector.

Figure 2. Comparison of spectral analyses of enriched ^{184}Os and background counting samples.

The value obtained for the counting efficiency was $36.4 \pm 3.6\%$, the error being compounded from the uncertainty of the ^{210}Po calibration and an estimated error of 5% introduced by scattering effects.

From this counting efficiency and from the upper limit of the ^{184}Os net counting rate, the maximum disintegration rate of the enriched ^{184}Os sample is calculated to be 0.0135 dpm. Using Viola, et al's upper limit of 3.1×10^{13} years for the half-life of ^{184}Os [V. E. Viola, Jr., C. T. Roche and M. M. Minor, J. Inorg. Nucl. Chem. 36, (in press, 1974)] the minimum disintegration rate which can be ascribed to the excess of ^{184}Os atoms in the sample over those in the blank is obtained as 0.0002 dpm. This disintegration rate constitutes only a small contribution relative to the statistical interval containing the "true" counting rate of the ^{184}Os sample; it was thus subtracted from the maximum disintegration rate of the sample, giving a maximum disintegration rate ascribable to the excess of ^{184}Os atoms in the sample over those in the blank of 0.0133 dpm. From this disintegration rate we obtain a lower limit of 4.3×10^{13} years for the alpha decay half-life of ^{184}Os . This work is currently being prepared for publication.

C. Radioactivity in Mollusc Shells

Our preliminary study of the incorporation of ^{90}Sr into the shells of certain species of marine bivalves from Venezuelan coastal waters was begun and completed during 1974.

This study was undertaken partly to establish baseline levels of artificial radioactivity in an area which has not as yet begun to develop nuclear power facilities. Also, since previous studies of the possible ecological effects of a nuclear-excavated sea-level canal through the Isthmus of Panama did not include the Venezuelan coast, which is at the latitude of and eastward of some of the suggested routes for the canal, it was felt that marine radioecological information in this region would be valuable, especially since seashells appear to offer a biological monitoring mechanism for fission products in the sea. Third, little appears to be known in general about the incorporation of strontium into molluscan shells or, indeed, about the total rates of growth of these shells. Careful radiostrontium studies by radiochemists can possibly contribute to this area of marine biology which is of very practical concern in the increasing number of maricultural efforts to "farm" edible molluscs such as clams, oysters and mussels.

Accordingly, a preliminary development of methods for radiostrontium assay in shell material has been accomplished, and specimens of two species of bivalves have been assayed. The specimens

were collected alive in February of 1974 in Mochima Bay, State of Sucre, Venezuela. Mochima Bay is located on the Caribbean coast in eastern Venezuela, between Puerto LaCruz and Cumaná, at a latitude of 10 to 11°N. The two species were the cross-barred venus clam (*Chione cancellata*) and the rough lima (*Lima scabra*). The shells were separated from the soft parts, washed in fresh water, dried in the sun and kept in sealed plastic bags until they could be analyzed. Four specimens of each species were ashed in a muffle furnace at 500°C. One valve of each specimen was pulverized in a mortar, dried for 15 minutes at 100°C and then sampled. One or two gram samples were taken for each determination.

A radiochemical separation procedure for strontium was devised. Yttrium-90 was milked from the purified strontium fractions and counted to determine the ^{90}Sr activities. Within statistical error, no activity was found in either group of samples, which sets an upper limit of 0.02 dpm ^{90}Sr /g ashed shell. This may be compared with a value of 16 dpm/g ashed shell found in giant clam shell material (*Tridacna gigas*) at Bikini Atoll [K. Bonham, Science 149, 300 (1965)] and a value of 5.8 dpm/g ashed shell found in upper Mississippi River clam shells [G. Pahl, Proc. Second Natnl. Symp. Radioecology, May 15-17, 1967, Ann Arbor, Mich. Conf. 670503, p. 234 (1969)].

Because different species contain different amounts of strontium, the significant quantity for contamination comparisons is the activity of radiostrontium per milligram of total strontium;

a method for the determination of total strontium was therefore devised. Because of complications caused by the large amounts of calcium present in the shell samples, total strontium was determined by an isotope dilution method using ^{86}Sr . The total strontium analyses gave results of 3.7 ± 0.2 mg Sr/g ashed shell in four specimens of *Chione cancellata* and 5.8 ± 0.6 mg Sr/g ashed shell in four specimens of *Lima scabra*. Many more analyses, of course, are needed to determine whether this is a real interspecies difference. However, these results can be used to calculate strontium concentration factors, the factors by which the molluscs' mantles concentrate the element strontium from sea water. Using the accepted average value of 8 mg Sr per liter of sea water, the concentration factors (mg Sr/g ashed shell $\div$ mg Sr/l sea water) are about 700 and 500 for *L. scabra* and *C. cancellata*, respectively. Using these concentration factors and the upper limit found for the ^{90}Sr activity in the shells and assuming no isotopic differences between the molluscs' utilization of stable and radiostrontium, we obtain an upper limit for ^{90}Sr activity in Mochima Bay water of 0.01 - 0.02 pCi/l. This may be compared with a direct measurement made in 1960 of 0.07 pCi/l in water from the open Caribbean sea. [I. Mauchline and W. L. Templeton, *Oceanog. Mar. Biol. Ann. Rev.* 2, 229 (1964)]. While no conclusions can be drawn from this single preliminary result, studies of this type clearly hold implications regarding fallout contamination from atmospheric nuclear explosions since 1960, and regarding the exchange of water between Mochima Bay and the open sea.

D. Review of Applications of Radiochemical Methods
in Oceanographic Research

A comprehensive literature review was prepared of the uses of radiochemical techniques in oceanography. The objective of this survey (which constituted the M.S. thesis of a graduate student) was to try to identify areas in which our low-level counting facilities could be useful and, more generally, to identify areas in which the skills of trained radiochemists could complement those of marine scientists to the enhancement of oceanographic knowledge.

To satisfy the purposes of the present report, we reproduce here only the table of contents of that review.

I. INTRODUCTION

- A. Oceanography
- B. Chemical Oceanography and Marine Chemistry

II. RADIOACTIVITY IN THE OCEANS

- A. Natural Radioactivity
- B. Artificial Radioactivity
- C. Analysis of Radioactivity in the Oceans
 - 1. Analysis of Radioactivity in Sea Water
 - 2. Analysis of Radioactivity in Sea Sediments
- D. Uptake of Radioisotopes by Marine Organisms
 - 1. Uptake of ^{90}Sr by Marine Organisms
 - 2. Uptake of ^{65}Zn and other Radionuclides by Marine Organisms

III. APPLICATIONS OF RADIOISOTOPES IN OCEANOGRAPHY

A. Dating with Radioactive Elements

1. Radiocarbon Dating
2. Other Cosmic-ray Induced Radionuclides Used in Dating
3. Uranium Decay Series
4. Potassium-Argon Dating

B. Determination of Sediment Transport and Density with Radioactive Tracers

1. Sediment Transport Determination
2. Sediment Density Determination

C. Oceanic Mixing and Circulation Studies with radioactive Tracers

1. Carbon-14 as Tracer for Oceanic Mixing
2. Tritium as Tracer for Oceanic Mixing
3. Other Radioactive Tracers for Oceanic Mixing

D. Determination of Primary Productivity of the Oceans Using ^{14}C as Tracer

E. Application of the Uptake of Radioisotopes by Marine Organisms

1. Applications of Radioactive Uptake to the Study of Marine Organisms Themselves
2. Use of Marine Organisms as Biological Monitors of Radioactivity

F. Analysis of Trace Elements in the Oceans by Neutron Activation

IV. CONCLUSIONS AND SUGGESTIONS

V. BIBLIOGRAPHY (Approximately 225 references)

E. The Statistics of Low-Level Counting

Experience gained in several of our studies involving environmental-level, background-dominant radioactivity measurements (e.g. Sections B and C of this report and previous work) has pointed out certain deficiencies in the standard treatments of the statistical aspects of radioactivity measurements. The treatments contained in the common radiochemical texts and monographs, while quite adequate for the counting of samples in which the background rate is relatively small, fail to consider those aspects of statistical theory which become quite important in the design of measurements and the interpretation of results in highly background-dominant situations. Moreover, what literature does exist on background-dominant counting is replete with inconsistent notation and, in some cases, statistically indefensible assumptions.

As a result of this realization, a critical examination of the radioactivity measurement process in this region has been undertaken during the past year. After development of a suitable descriptive model of the measurement process, interest was focused on a function which we call the "minimum probability of detection", which is related to the power function in the classical statistical theory of hypothesis-testing. This function has the useful advantage of containing only observed quantities, and it allows one to formulate a criterion for whether or not the data permit one to deduce a specific minimum probability of detection. However, as an a posteriori

quantity, it does not provide information which can be used for the choice of experimental parameters such as counting time, total number of counts, etc. To this end, an a priori approximation was developed which allows prior estimation of the values of these quantities which are necessary to achieve a given minimum probability of detection. At present, the validity of this approximation in various circumstances is being investigated.

The initial hypothesis-testing problem of whether or not a sample contains a net activity above background has been generalized to the question of whether or not a sample exceeds some specified maximum permissible level of activity. This question is of great importance, for example, in the monitoring of effluents from nuclear activities. Here again, both an a posteriori criterion and an a priori approximation are formulated.

At present, the work is being cast into a form which would be suitable for practical everyday use at the technician's level.

F. Computation

The on-line computer facility at the Nuclear Physics Laboratory has been expanded this year through the acquisition of a second PDP-15 computer. The two computers are presently linked by 8K words of shared memory. This permits the fast transfer of data between the two machines and makes available to either computer any of the I/O peripheral devices.

On-line data acquisition is controlled by the Nuclear Physics Laboratory codes SNAPPY and CRUNCH. SNAPPY is used to obtain singles data and CRUNCH to obtain coincidence data. A data analysis program is now being written which, in conjunction with CRUNCH, will permit complete on-line analysis of a charge-state experiment.

Our use of the two PDP-10 computers operated by the University of Pittsburgh's Computer center continued to be extensive in 1974. Several new utility programs were written, including plotting routines, linear least squares routines, and a routine to invert Laplace transforms numerically. To facilitate the execution of PDP-10 programs, a Porta-Com computer terminal and telephone coupler were obtained. This portable terminal allows us to access either of the PDP-10's directly from our laboratory.

G. New Facilities and Equipment

1. New Laboratories.

At the time of last year's report, the Low Beta II low-level counter had been moved into our counting room in the new chemistry building several months before the Chemistry Department, including the rest of our group's activities, was to move in. This was done so that the low-level ^{90}Sr and ^{137}Cs counting could be done in an uninterrupted sequence, in completely uncontaminated surroundings. During the year, our entire program has moved into the new building. The Wherrett Laboratories now occupy some 2700 square feet of space on the eighth floor of this modern fifteen-story building, plus an isotope storage facility on the first floor. The eighth-floor space consists of a laboratory for medium-to-high-level radiochemical work which doubles as a teaching laboratory in radiochemical methods one semester per year, a low-to-medium-level lab, an instrument room and target-preparation lab, a calculation room and graduate student office, a counting room which doubles for research and teaching, and office space for the Principal Investigator and secretary.

2. Negative Ion Source

A sputter-type ion source has been installed in the Van de Graaff. This source, an Extrion UNIS (Universal Negative Ion Source), is capable of producing a wide variety of elemental ion beams at high intensity. It produces negative ions by a sputtering process with cesium. The installation of this source has permitted us to begin

the heavy ion charge-state program reported in Section A of this report.

3. Evaporator System

For years, our nuclear chemistry group has shared the Nuclear Physics Lab's heavily-used vacuum evaporation system for preparing Van de Graaff targets. In 1974, using funds from the Pennsylvania Department of Property and Supplies' allocation for equipping the new chemistry building, we acquired our own Varian NRC 3117 Vacuum Coater System. This system is equipped with a six-inch 1500 l/sec diffusion pump with a liquid nitrogen cryobaffle, a 10.6 cfm mechanical pump, self-contained circuitry for the operation of vacuum gauges, and an 18" X 30" Pyrex bell jar with implosion shield. To permit rapid changeover of vacuum feedthroughs, an 18" stainless steel feedthrough collar was added to the basic system. The system is equipped with a built-in 2 kVa resistance-heating power supply and an electron beam gun. The electron gun is capable of heating samples to temperatures in excess of 3500°C with an electron beam output of 0-500 mA at 4 kV. The electron beam impingement position can be adjusted over a range of about 1/8 inch at full power.

4. Counting Equipment

Most of the electronics in our counting room at present are old vacuum-tube circuits acquired some thirteen years ago. Using the Pennsylvania Department of Property and Supplies' new building equipment fund, we have ordered modern NIM electronics to update most of the detection equipment in the counting room. This combined research and teaching facility contains a Beckman Low-Beta II low-level

counter, eight geigercounters, two proportional flow counters, a 4π proportional counter, a three-inch sodium iodide crystal with 512-channel pulse-height analyzer, a two-inch sodium iodide well crystal, and a liquid scintillation spectrometer with 200-channel pulse-height analyzer. . New NIM solid-state electronics are being provided for the geiger, proportional and NaI detectors.

III. PERSONNEL

Principal Investigator

Robert L. Wolke, Professor of Chemistry. B.S. Chem. Polytechnic Institute of Brooklyn 1949, Ph.D. Cornell University 1953. (Approximately one-third time January 1974 through April 1974, approximately one-third time May 1974 through December 1974.)

Other Research Personnel

Ali Hussain Baha Al-Deen, Professor of Chemistry, Universidad de Oriente, Cumaná, Venezuela. Venezuelan Fellow. (Not supported on AEC contract funds). Diploma in Engineering, University of Zagreb, Yugoslavia. M.S. University of Pittsburgh, completed April 1974. (Radioactivity in Mollusc Shells)

Biserka Krog de Baha Al-Deen, Professor of Chemistry, Universidad de Oriente, Cumaná, Venezuela. Venezuelan Fellow. (Not supported on AEC contract funds). Diploma in Engineering, University of Zagreb, Yugoslavia. M.S. University of Pittsburgh, completed April 1974. (Radiochemical Methods in Oceanography)

Judith C. Brillhart, Graduate Research Assistant July 1974 - September 1974. B.S. Western Illinois University 1971. (Radioactivity in Mollusc Shells)

Jack J. Donn, Captain, U. S. Air Force. Civilian Institutions Student, U.S. Air Force Institute of Technology (Not supported on AEC contract funds.) B.S. University of Cincinnati 1968. (Statistics of Low-Level Counting)

William J. Jordan, Mellon Fellow. (Not supported on AEC contract funds.) B.S. University of Pittsburgh 1973. (Charge States of Heavy Ions and Heavy Ion Nuclear Reactions)

Robert F. Sperlein, Lieutenant, U.S. Air Force. Civilian Institutions Student, Air Force Institute of Technology. (Not supported on AEC contract funds.) B.S. University of Pittsburgh, 1973. M.S. University of Pittsburgh, completed August 1974. (Natural Radioactivity in Osmium)

John D. Yesso, Graduate Research Assistant January 1974 to present. B.S. University of Pittsburgh 1971. (Charge States of Heavy Ions)

Other Personnel

Patricia L. Moan, Secretary. April 1974 to present.

IV. PUBLICATIONS AND TALKS

(Publications listed have appeared in 1974, have been submitted for publication, or are scheduled for submission before the end of the current contract year.)

Charge-Changing Cross Sections in Boron of 7-MeV Boron-11 Recoils from the Elastic Scattering of Deuterons. E. V. Mason, Jr., R. L. Wolke, T. W. Debiak and J. D. Yesso, Phys. Rev. A 9, 1569 (1974).

Spin Assignments in the Decay of Indium-109. S. Shastry, T. W. Debiak, E. V. Mason, Jr., J. D. Yesso and R. L. Wolke, (Submitted to Particles and Nuclei)

Charge-Changing Cross Sections in Carbon of ^{10}B and ^{12}C Recoils from Nuclear Reactions. R. L. Wolke, E. V. Mason, Jr., R. A. Sorbo, T. W. Debiak and J. D. Yesso. (In preparation)

Charge-Changing Cross Sections of Lithium, Beryllium and Boron Ions Produced as Recoils from Nuclear Processes. T. W. Debiak, R. L. Wolke, J. D. Yesso and W. J. Jordan. (In preparation)

A Search for Alpha Instability in Osmium-184. Robert F. Sperlein and Robert L. Wolke (In preparation)

R. L. Wolke, "Problemas de la Radiactividad en el Medio Ambiente" Graduate seminar, Oceanographic Institute, Universidad de Oriente, Cumaná, Venezuela. 7 February 1974

R. L. Wolke, "Low Level Radioactivity in Isobars and Sandbars." Seminar, Puerto Rico Nuclear Center, Mayagüez, P. R. 10 June 1974.

R. L. Wolke, "The Nuclear Chemistry Research Program at Pitt." Graduate research seminar, Department of Chemistry, University of Pittsburgh 7 November 1974.

(Plus other invited talks on topics in chemical education.)